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SOOT MORPHOLOGY IN UNSTEADY COUNTERFLOW DIFFUSION FLAMES

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SUMMARY/OVERVIEW:

As Diesel engines consume the majority of the injected fuel in diffusion controlled combustion processes compared to the relatively short initial premixed phase, and diffusion flames have a greater propensity to form soot, particulate matter emission from Diesel engines can considerable. These particles have a much stronger thermal signature compared to gas phase products, water and carbon dioxide, and render Diesel-powered vehicles susceptible to tracking and targeting via IR sensors. This IR signature will decay with time as the particles cool, and this temporal profile is a function of the morphology of the soot. Therefore, it is important to understand, and eventually control, not only the soot volume fraction of the particulate matter, but also its morphology.

TECHNICAL DISCUSSION:

In order to understand the very complicated coupling between the three dimensional, unsteady fluid dynamics in a turbulent flowfield and the chemical kinetics of combustion, it is necessary to simplify either the flowfield or the chemistry, or both. The turbulent flame has been successfully modeled as an ensemble of one-dimensional strained laminar flamelets under certain conditions [1]. A counterflow geometry as been used for many years as both a computational and experimental model of such a flamelet [2]. Until recently, the structure of these flamelets was assumed to be only a function of the strain rate, defined as the air side axial velocity gradient just prior to the heat release zone. However, it has become apparent that knowledge of the instantaneous strain rate is not sufficient, and the history must also be known for rapidly varying strain rates [3,4].

DeCroix and Roberts [5] have measured the soot volume fraction in an unsteady counterflow diffusion flame burner. Unsteadiness was imposed on the flame by oscillating the reactant flow rates, thereby imposing an oscillation in the strain rate. Measurements of f_{sv} were made as a function of initial strain rate, oscillation frequency and oscillation amplitude using a calibrated Laser Induced Incandescence (LII) technique. Table 1 shows results of these measurements, where A_1 and A_2 are two different forcing amplitudes, relative to the respective global quenching or flow reversal amplitude, which ever is less, at each initial strain rate. The

values in the table are normalized to the steady peak soot volume fraction measured at the corresponding initial strain rate. As seen in this table, at low frequencies, the unsteady flames have peak soot volume fractions that are as much as six times higher than their steady counterpart. The peak augmentation occurs for moderately low initial strain rates. Below this, the flame is already heavily sooting and the unsteadiness contributes relatively little. At higher oscillation frequencies, there can be a large difference in the behavior of the peak soot volume fraction, depending upon the initial strain rate. At low initial strain rates, the higher oscillation frequencies are seen to drastically reduce the peak f_{sv}, while at higher initial strain rates, the f_{sv} becomes relatively insensitive to the imposed oscillations.

Frequency	SR 15 s^{-1} Steady max f_{sv}=1.00 ppm		SR 30 s^{-1} Steady max. f_{sv}=0.20 ppm		SR 60 s^{-1} Steady max. f_{sv}=0.05 ppm		SR 90 s^{-1} Steady max. f_{sv}=0.03 ppm	
(Hz)	A$_1$	A$_2$	A$_1$	A$_2$	A$_1$	A$_2$	A$_1$	A$_2$
25	2.4	1.9	3.4	6.5	1.8	3.4	2.0	3.0
50	1.1	0.7	1.5	1.9	1.6	1.4	1.7	2.3
100	0.9	0.3	1.3	0.9	~1	~1	~1	~1
200	0.8	0.1	1.1	0.5	~1	~1	~1	~1

Table 1 Peak soot volume fraction in unsteady counterflow diffusion flame burner as a function of initial strain rate, oscillation frequency and amplitude, as measured with LII.

As stated above, the morphology of the soot is of importance in determining the radiation signature. LII has been very successful at measuring the f_{sv}, but there are difficulties in deriving particle size information from the incandescence signal. As the flow field is sensitive to physical obstructions, sampling probes are problematic. In-situ light scattering and extinction measurements, therefore, have usually been employed in order to obtain critical soot data in flames. Almost all of the early literature inferred soot particle size and number densities using Mie or Raleigh theories [6,7]. Typical soot primary particle size is 30 – 50 nm, much smaller then the wavelength of visible radiation, and fully within the Rayleigh regime. However, soot is composed of aggregates containing hundreds to thousands of primary particles. The effective diameter of these aggregates is considerably larger than the wavelength of visible radiation, fully within the Mie regime. Unfortunately, these primary particles do not cluster into a larger sphere, rendering both scattering models inaccurate, but not irrelevant [8,9].

By making a single extinction measurement along with angularly resolving the scattered light, looking at all four polarization options, the morphology of the soot may be determined. The main difficulty in the interpretation of these optical measurements was a scattering theory that relates optical cross section to aggregate morphology and size. This obstacle has been overcome by recent developments in fractal concepts. RDG/PFA theory [10] was found to be a reliable approximation to evaluate the measured optical cross sections of soot aggregates, based on extensive experimental [11,12] and computational [13] evaluations. RDG/PFA provides a general approach that yields N$_g$ (geometric mean of number of primary particles per aggregate) and σ_g (for a log-normal distribution), and fractal prefactor k and mass fractal dimension, D$_f$.

This theory also allows determination of the probability density function of N as well as the primary particle diameter, which are the most crucial parameters in particle growth and aggregation studies.

This approach requires numerous angular measurements, and may not be practical in many environments. By making a few experimentally-justified assumptions, the number of angular measurements can be reduced significantly. It has been shown that the fractal prefactor and fractal dimension of the soot aggregate are fairly constant for a wide range of fuels and flame geometries [14,15]. Also, the number of spherules per aggregate does follow a log-normal distribution whose width is fairly constant. Thus, by assuming the k, D_f, and σ_g are known, the number of angular measurements can be reduced to three.

Thus, by making a few good assumptions, RDG/PFA theory may be used to obtain soot morphological parameters in the unsteady counterflow diffusion flame burner as a function of steady strain rate and oscillation frequency and amplitude. Currently, this technique has only been used at a point using a focused cw laser and photomultiplier tube. However, if the focused beam is replaced with a sheet and the pmt is replaced with an ICCD camera, planar measurements of soot morphology may be possible. Sufficient laser fluence is necessary to obtain measurable signals, especially normal to the laser beam. However, if the fluence is too high, then unwanted photons from laser induced incandescence and laser induced fluorescence processes become a significant problem. Therefore, an un-Q-switched Nd:YAG will be used, as shown in Figure 1 below.
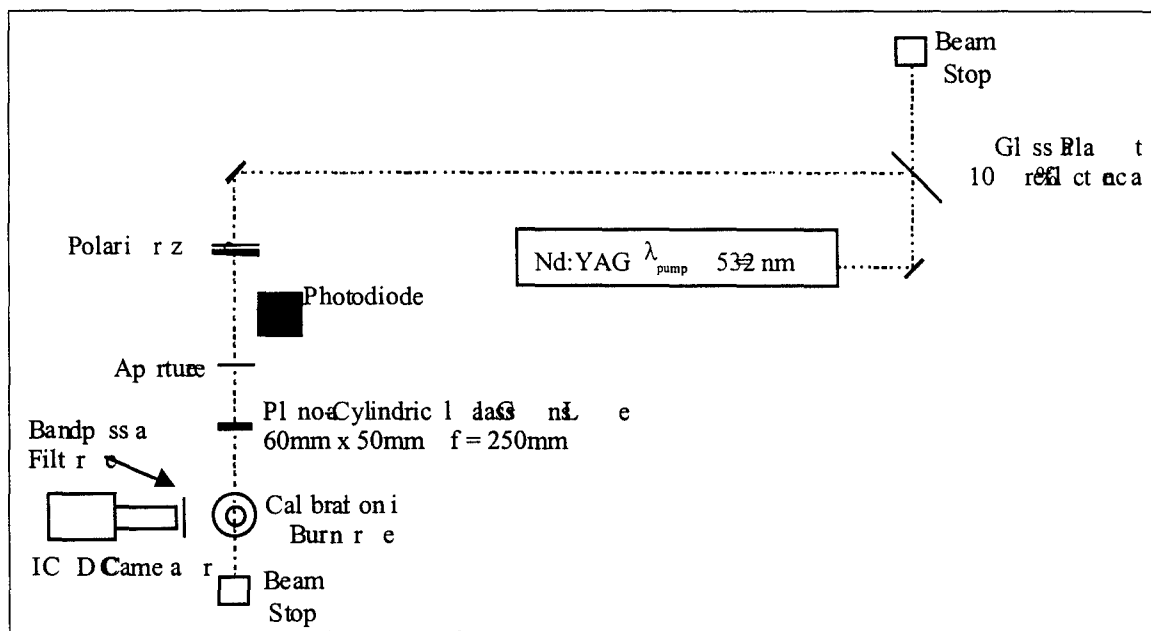

Figure 1. Planar RDG/PFA soot morphology measurement layout.

Currently, the PI is making point measurements in a laminar co-flow diffusion flame, identical to the "Santoro" burner, which is extensively characterized in the literature. Upon completion of these point measurements, planar measurements in this same flame will begin. By

developing this planar RDG/PFA technique in a well characterized, steady, axisymmetric flame, correction algorithms for extending the point technique to a planar technique can be devised and tested. When spatially resolved soot morphology measurements are completed in the co-flow flame, the "Santoro" burner will be replaced with the unsteady counterflow diffusion flame burner.

Soot morphology measurements will be made at the same conditions as listed in table 1. Concurrently, the temperature field is also being measured in this burner [16] at these same locations. Upon completion of these two studies, the effects of unsteady strain rate on the formation of soot and its morphology will be well quantified in these one dimensional flamelets.

References:

1. Peters, N. (1984). *Prog. Energy Comb. Sci.*, Vol. 10, pp. 319-339.
2. Tsuji, H. (1982). *Prog. Energy Comb. Sci.*, Vol. 8, pp. 93-119.
3. Im, H. G., Law, C. K., Kim, J. S., Williams, F.A., *Combust. Flame* **100**, pp. 21-30 (1995).
4. Egolfopoulos, F. N., Campbell, C. S., *J. Fluid Mech.* **318**, pp. 1-29 (1996).
5. DeCroix, M. E., Roberts, W. L., *Comb. Sci. and Tech* **160**, pp. 165-190 (2000).
6. Kent, J.H. and Wagner, H.Gg. (1982). *Comb. Flame*, **47**, 53-65.
7. Santoro, R. J., Semerjian, H.G., and Dobbins, R.A. (1983). *Comb. Flame*, **51**:203-218.
8. Köylü, Ü. Ö., and Faeth, G. M. (1993). *J. Heat Transfer*, **115**:409-417.
9. Köylü, Ü. Ö., and Faeth, G. M., (1996). *J. Heat Transfer*, **118**:415-421.
10. Moutain, R.D., and Mulholland, G.W., (1988). *Langmuir*-4:1321-1326.
11. Köylü, Ü. Ö., and Faeth, G. M., (1994a). *J. Heat Transfer,* **115**:971-979.
12. Köylü, Ü. Ö., and Faeth, G. M., (1994b). *J. Heat Transfer*, **116**:152-159.
13. Farias. T.L., Carvalho, M.G., Köylü, Ü. Ö., and Faeth, G. M., (1995). *J. Heat Transfer*, **117**:152-159.
14. De Iuliis, S., Cignoli, F., Benecchi, S., and Zizak, G., Proc. Comb. Inst, Vol. 27, pp. 1549-1555 (1998)
15. De Iuliis, S., Cignoli, F., Benecchi, S., and Zizak, G., Applied Optics, Vol. 37, No. 33 (1998)
16. Welle, E. J., Roberts, W. L., Donbar, J. M., Carter, C.D., DeCroix, M.E., Proc. Combust. Inst. 28, (2000).